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SLOPE–SLOPE CORRELATIONS FOR SELF-AFFINE ROUGH SURFACES

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The slope–slope correlation function $N(r)$ is investigated for self-affine rough surfaces. Calculations of $N(r)$ are performed in terms of analytic phenomenological height–height correlation functions, which however compare well to real data. It is found that $N(r)$ behaves as: $N(r) \propto r^{2(H-1)}$ for $0 \ll r \ll \xi$, $N(r) < 0$ for $r > \xi$ and $N(r) \to 0^-$ for $r \to +\infty$. The parameters ξ and H $(0 < H < 1)$ are respectively the in-plene roughness correlation length, and the roughness exponent. Moreover, connection of the results to model predictions describing stable and unstable growth is attempted. Copyright © 1996 Published by Elsevier Science Ltd

Orientation correlation functions have been widely used to describe the fluctuations of *membrane*-like surfaces [1] and surfaces grown under non-equilibrium conditions [2, 3]. In the latter case, for growth dominated by surface diffusion in the harmonic approximation (absence of non-linearities), the orientation correlation function scales as $\sim \ln(r)$ (r is the in-plane position vector) in $(2+1)$-dimensions [2]. The presence of non-linearities during growth (depending on the dimensionality) can cause power law scaling of the orientation correlation functions [2].

Furthermore, during growth by means of Molecular Beam Epitaxy (MBE) in the presence of step-edge (Schwoebel) barriers, there is selection of a critical slope ($m = |\nabla z(r)|$, where $z(r)$ is the surface height and the in-plane position vector) that leads to unstable growth with the formation of large-scale mounds over a singular surface (i.e. a low index crystal face), or to stable growth of a vicinal surface with a miscut above a certain value [3]. In fact, growth instabilities manifest themselves as pyramid-like structures [4]. It has been suggested that the classification of the growth law can be determined from the temporal evolution of the first zero of the slope–slope correlation function $N(r) \sim \Sigma_{\{p\}} < m(p)m(r+p) >$ [4]. This functional was also used to determine the scaling behaviour of the domain size in the *XY*-model [5].

The non-equilibrium process besides the instabilities, however, leads in many cases to scale invariance of the correlation functions which is manifested as self-affine fractal scaling [6]. Natural examples of self-affine topology are the nanometer scale topology of vapour deposited films [7], the spatial fluctuations of liquid–gas interfaces [8], the kilometer-scale structures of mountain terrain [6], etc. Physical processes which produce such a topology are fracture, erosion and MBE, as well as fluid invasion of porous media [9].

A thorough study of properties of the slope–slope correlations for self-affine fractals is still missing. In our study, we shall examine properties of slope–slope correlations in terms of phenomenological height–height correlation models which, however, can describe real data significantly well.

All rough surfaces exhibit perpendicular fluctuations which are characterised by a mean-square roughness $\sigma = < [z(r)]^2 >^{1/2}$; $< z(r) > = 0$, where $< \dots >$ is an average over the whole planar reference surface. For an isotropic rough surface, the height-difference correlation function $g(r)$ is written as $g(r) = < [z(r) - z(0)]^2 >$. For any physical self-affine surface $g(r)$ will saturate at large length scales to the value $2\sigma^2$. Thus, it is characterised by a finite correlation length ξ [10, 11] such that $g(r) \sim r^{2H}$ if $r \ll \xi$ and $g(r) = 2\sigma^2$ if $r \gg \xi$. $H (0 < H < 1)$ is the roughness exponent which characterises the degree of surface irregularity [12]. Small values of $H \sim 0$ correspond to extremely jagged or irregular surfaces, while large values of $H \sim 1$ to surfaces with smooth hills and valleys [10]. The function $g(r)$ is related to the height–height correlation function $C(r) = < z(r)z(0) >$ by means of $g(r) = 2\sigma^2 - 2C(r)$.

As mentioned earlier, the fluctuations of *membrane*

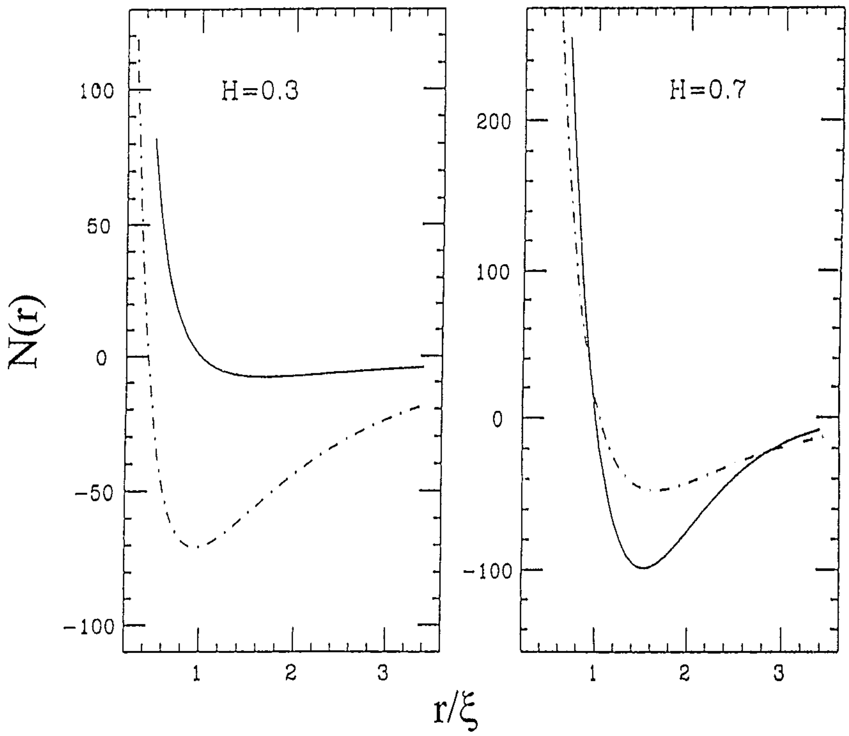

Fig. 1. Schematics for the slope–slope correlation with $\sigma = 0.7$ nm, and $\xi = 100$ nm. $N_s(r)$ solid line; $N_k(r)$, dot–dashes.

surfaces are more conveniently characterised by the orientation correlation function $G(r) = \langle [\nabla z(r'') - \nabla z(r')]^2 \rangle (r = r'' - r')$ [1, 2]. The *slope–slope* correlation function $N(r) = \langle \nabla z(r'') \nabla z(r') \rangle$ is related to $G(r)$ by means of $G(r) = 2 \langle [\nabla z(r)]^2 \rangle - 2N(r)$. For an orientationally disordered surface $G(\infty)$ diverges, while for an asymptotically flat surface $G(\infty)$ is a constant [1, 2]. Let us denote the average macroscopic sample surface by A. We define $C(r)$ by $C(r) = 1/A \int \langle z(p+r)z(p) \rangle \mathrm{d}^2p$, and $N(r)$ by $N(r) = 1/A \int \langle \nabla z(p+r) \nabla z(p) \rangle \mathrm{d}^2p$. The symbol $\langle \ldots \rangle$ denotes an ensemble average over possible roughness configurations. A simple relation exists in between $C(r)$ and $N(r)$, namely $N(r) = -\nabla^2 C(r)$. Alternatively for isotropic surfaces, if we define by $C(k)$ the Fourier transform of $C(r)[C(r) = \int C(k) \mathrm{e}^{ikr} \mathrm{d}^2k]$, we obtain

$$N(r) = -\nabla^2 C(r) \quad \Leftrightarrow \quad N(r) = 2\pi \int_0^{kc} k^3 C(k) J_0(kr) \mathrm{d}k, \tag{1}$$

where $J_0(x)$ is the zero-order Bessel function. $k_c = \pi/a_0$ with a_0 the atomic spacing. The upper cut-off is related with the fact that any notion of fractal scaling at length scales below a_0 ceases to exist.

We will perform explicit calculations of $N(r)$ for two correlation models. The first one is the so-called *stretched* exponential [11, 13] $C_s(r) = \sigma^2 \mathrm{e}^{-(r/\xi)^{2H}}$, which has been used in a wide range of relaxation phenomena studies [13] and can be utilised to study the limiting case $H = 1$ (observed in unstable MBE-growth of pyramid-like structures) explicitly. The second model is the so-called *k-correlation* function which has an analytic Fourier transform $C_k(k) = (\sigma^2 \xi^2 / 2\pi)[1 + ak^2\xi^2]^{-1-H}$ with $a = (1/2H)[1 - (1 + ak_c^2\xi^2)^{-H}]$ if $0 < H < 1$ and $a = 1/2 \ln(1 + ak_c^2\xi^2)$ if $H = 0$. This model can be used also to investigate the case of logarithmic roughness ($H = 0$) as a limiting case of self-affine roughness [14]. The logarithmic roughness ($H = 0$) is related to predictions of various growth models of the non-equilibrium analogue [15] of the equilibrium roughening transition [16].

In both cases from equation (1), we obtain

$$N_s(r) = \frac{(2H)^2}{\xi^2} C_s(r) \left(\frac{r}{\xi}\right)^{2(H-1)} \left[1 - \left(\frac{r}{\xi}\right)^{2H}\right];$$

$$N_k(r) = \sigma^2 \xi^2 \int_0^{k_c} \frac{k^3 J_0(kr)}{(1 + ak^2\xi^2)^{1+H}} \mathrm{d}k, \tag{2}$$

$$N_k(r) = \frac{\sigma^2 \xi^2}{a^2 \Gamma(1+H)} \left(\frac{r}{2a^{1/2}\xi}\right)^H K_{2-H}\left(\frac{r}{2a^{1/2}\xi}\right)$$

$$\times (H > 1/4\Lambda r, \xi \gg a_0),$$

where $k_{2-H}(x)$ is the $(2-H)$-order second kind Bessel function.

Plots of $N_{s,k}(r)$ are shown in Fig. 1, where it can be observed that the slope–slope correlation function reveals significant structure in the regime of length scales $r \sim \xi$ (minimum). This is explained by the fact that for length scales $r < \sim\xi$ the slopes repel each other leading to disorder and for $r > \xi$ are attracted leading to an ordered structure in such a way that the surface is asymptotically flat (for $r \gg \xi$). The depth of the well for $N_k(r)$ (Fig. 1) increases as H decreases, which implies that the smaller the H the larger is the attraction of the slopes for length scales $\sim\xi$. For $N_s(r)$, the apparent opposite behaviour as a function of H is related with the fact that the correlation function $C_s(r)$ inverts its decay rate at $r \gtrsim \xi$ (see first of [13]) and for small $H (H \sim 0)$ shows a completely unphysical behaviour [14]. The minimum ($R_{\min}$) is a function of H and ξ, and increases with increasing H. In fact, for $Ns(r)$ is given explicitly by $R_{\min}/\xi = [(6H - 2 + [20H^2 - 8H + 4])/2H]^{1/2H}$.

Furthermore, we have $N_s(0, H < 1) = \infty$ and $N_s(0, H = 1) = 4\sigma^2/\xi^2$. Moreover, at $a_0 \ll r \ll \xi$ we have $N_s(r) \sim (r/\xi)^{2(H-1)}$. On the other hand for $N_k(r)$, we have $N_k(0) = (\sigma^2\xi^2/2a^2)\{[1/(1-H)]\ [1 + akc^2\ \xi^2)^{1-H} - 1] - 2a\}$ if $0 \le H < 1$ and $N_k(0) = (\sigma^2\xi^2/2a^2)$ $\{\ln(1 + ak_c^2\xi^2) - 2a\}$ if $H = 1$. For $a_0 \ll r \ll \xi$ we have $N_k(r) \sim (r/\xi)^{2(H-1)}$ and for $r \gg \xi$ we have the asymptotic behaviour $N_k(r) \sim (r/\xi)^{H-1/2} \exp(-r/\xi)$. Therefore from the previous, the general behaviour of $N(r)$ for $0 \le H < 1$ can be described briefly from the following scheme.

$$N(r) \propto \begin{matrix} \sigma^2/\xi^2 \text{ if } r \to 0; & r^{2(H-1)} \text{ if } r \ll \xi \\ < 0 \text{ if } \xi < r < +\infty; & \to 0^- \text{ if } r \to +\infty. \end{matrix} \quad (3)$$

The apparent divergence at $r \sim 0$ is a result of the non-existence of a lower continuum limit which for the case of $N_k(r)$ has been taken into account and from the fact that $H < 1$. Scaling behaviour similar to power law $\sim r^{2(H-1)}$ has also been observed for $G(r)$ in studies of surface diffusion models (where depending on the dimensionality) we have; $G(r) \sim r^{2(H-1)}$ [2].

In Fig. 2, we present $N(r)$ Scanning-Tunneling-Microscopy (STM) data from an Ag-film deposited (at rate $0.03\,\text{nm}\,\text{s}^{-1}$) by means of thermal evaporation on a Quartz crystal substrate kept at room temperature (~300 K). The topographic images acquired with scan size 500 nm ($\gg\xi$). The data are averages from four STM images acquired on different surface locations. The inset shows the corresponding height–height correlation function which is well fitted by the $C_s(x)$ function for $r \le 2\xi$ with $\xi = 12.3$ nm and $H = 0.72$ (first of [13]).

The existence of a zero (R) of $N(r)$ [$N(R) = 0$] occurs for length scales $\sim\xi$ (see Fig. 1) with an exact value that depends on the particular model under investigation. In fact, for $N_s(r)$ the zero occurs exactly at $R = \xi$ independently of the value of H. While for $N_k(r)$ as H decreases, R also decreases. Nevertheless, it should also be pointed out that the slope-slope correlation function possesses *only* one zero which is a function of H and ξ. This point is important since in the case of $H = 1$ (unstable growth) the growth law can be deduced from the temporal dependence of this zero; $R \sim t^{1/z}$ with $3 \le z \le 4$ [4].

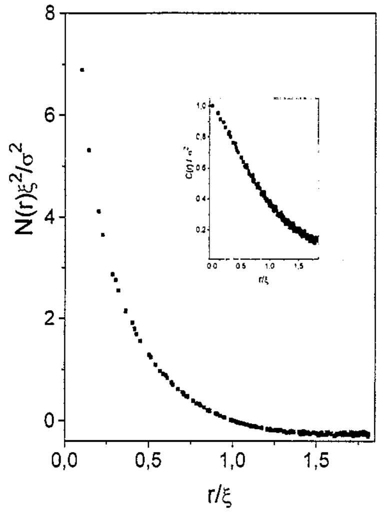

Fig. 2. $N(r)$-correlation data vs r/ξ for the room-temperature Ag-film with $H = 0.72$. The inset depicts the corresponding correlation data $C(r)$ vs r/ξ.

The formation of pyramid like structures observed in experimental studies of unstable growth possesses roughness exponent $H = 1$ [3, 4, 18]. The theory suggested recently by Siegert and Plischke [4] predicts that there is not any wavelength-selection: in an infinite system the pyramids would continue to grow, whereas in a finite system the surface profile saturates when only one pyramid remains. Similarly according to Johnson *et al.* [3], the mounds grow by keeping the angle the sloping sides make with the terraces constant, and further as the growth continues the mounds coalesce until only one of the order of the system size remains. At this point finite size effects cause the surface to saturate.

The case $H = 1$ can be treated in a simple way as a limiting case of self-affine structure by means of the correlation function $C_s(r) = \sigma^2 e^{-(r/\xi)^{2H}}$. In this case, $N_s(r, H = 1)$ is given by $N_s(r, H = 1) = 4(\sigma^2/\xi^2) e^{-(r/\xi)^2}\ [1 - (r/\xi)^2]$ with well-minimum at $R_{min} = 2^{1/2}\xi$ and zero at $R = \xi$. In fact the function $N_s(r, H = 1)$ suggests that the length scale ξ still remains the length scale, where its temporal evolution (i.e. by increasing film thickness during deposition) will define the growth law of the system [4]. Despite the fact that this conclusion is drawn from isotropic surfaces in the xy-plane, its validity is plausible to be extended to anisotropic surfaces.

Differences in the behaviour of the slope–slope correlations arise in (1 + 1)-dimensions. This can be seen easily if we calculate $N_s(x)$ from $C_s(x)$ correlation. From equation (1) we obtain $N_s(x) = (2H)^2(x/\xi)^{2H-2}$

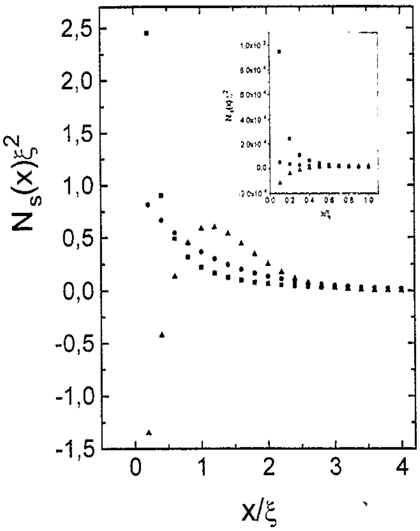

Fig. 3. Schematics for $N_s(x)$ vs x/ξ for $H = 0.3$: Squares, $H = 0.5$: Circles, $H = 0.8$: Up-triangle. The inset shows $N_1(x)$ vs x/ξ for $H = 0$, Squares; $H = 0.5$, Circles; $H = 1$, Up-triangle.

$[(x/\xi)^{2H} - (1 - 1/2H)]C_s(x)/\xi^2$ (see Fig. 3). We can observe that $N_s(x) > 0$ everywhere for $H < 1/2$, while $N_s(x) < 0$ for $H > 1/2$ in the regime of length scales $x < \xi(1/2H - 1)^{1/2H}$ and $N_s(x) > 0$ for $x > \xi(1/2H - 1)^{1/2H}$ (Fig. 3). However, in order to gauge the effect of model-dependence, we perform a calculation of the slope–slope correlation for another model, which is not problematic at very low roughness exponents $H(H \sim 0)$ as it happens for the $C_s(x)$ function [13].

Thus, we choose the model $C_1(k) = \sigma^2\xi(1 + a|k|\xi)^{-1-2H}$ [19] with the parameter "a" given by $a = (1/H)[1 - (1 + ak_c\xi)^{-2H}]$ $(0 < H \leq 1)$; $a = (1/2)\ln(1 + ak_c\xi)$ $(H = 0)$. The C_1-model has natural behaviour for all the exponents $0 \leq H \leq 1$ [20], where based on equation (1) we obtain $N_1(x) = 2\sigma^2\xi x^2 \int k^2(1 + ak\xi)^{-1-2H} dk$ $(0 \leq k \leq k_c)$. As can be seen in the inset of Fig. 3, we have similar behaviour with the $N_s(x)$ for large and small roughness exponents H. The only model dependence is on the value of H for which above that value [$H = 1/2$ for $N_s(x)$]$N(x) < 0$ at small length scales. Moreover, from both $(1 + 1)$-dimensional cases we infer that the existence of a zero [$N(x) = 0$] depends strongly on the value of H by contrast to $(2 + 1)$-dimensions. Therefore we can conclude that in $(1 + 1)$-dimensions, the slope-slope correlation posseses drastically different behaviour than in $(2 + 1)$-dimensions.

In conclusion, we investigated properties of the slope-slope correlation $N(r)$ for self-affine fractal morphologies. The degree of surface irregularity (which is depicted through H) plays a crucial role on the disordering-ordering process as a function of length scale, which is depicted by means of $N(r)$ (Fig. 1). Furthermore, our study revealed the existence of a unique zero of $N(r)$ for self-affine morphology, which can play an important role in determining the growth law in the limiting case of $H = 1$ (unstable growth) as has been already pointed out in previous studies [4]. Finally, drastic differences in the behaviour of the slope-slope correlation arise as a function of the surface imbedded space dimension.

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